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Consummate solution to the problem of classical electromagnetic scattering by an ensemble of spheres. II: Clusters of arbitrary configuration

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The order-of-scattering approach developed earlier [Opt. Lett. 13, 90 (1988)] and applied there to the case of linear chains of spheres is extended to the more difficult problem of scattering by clusters of spheres, the centers of which no longer need lie on a common axis. To help establish the validity of this most general calculation, comparisons are made between theoretical and experimental results for triangular and tetrahedral arrays of spheres. We also perform calculations based on an older method that requires the inversion of matrices, and we find that for the cases considered here the order-of-scattering method is substantially faster.

In a recent Letter[1] we reported on our development of an order-of-scattering (OS) technique that allows one to calculate the fields scattered by interacting spheres in complete detail. In that paper (referred to as Part I) we applied this new method to the special case of two or more spheres centered on a common axis. A theory based on matrix methods was developed in the late 1960's[2,3] that can also be used to determine the fields scattered by clusters of interacting spheres. This latter method was applied by its original authors to the case of linear chains made up of two or three spheres, the size parameters of which ranged from $ka \approx 4$ to $ka \approx 25$, where $ka = 2\pi a/\lambda$, with a the radius of a constituent sphere and λ the wavelength of the incident radiation in the medium surrounding the cluster. The intent of Part I was, in part, to validate the OS method by comparing the results of calculations based on both techniques, which were in turn compared with existing experimental data. Excellent agreement was obtained in such comparisons, and the OS technique was then applied to a study of the fields scattered by a linear chain of three spheres wherein the contributions to the scattered intensity by increasing orders of partial fields were also considered.

Computational demands made by their theory for clusters of more arbitrary morphologies seem to have prohibited Lo and his colleagues from applying that method to those systems. Such calculations are still rather taxing for moderate-sized modern computers. In this Letter we discuss how the OS method fares under such circumstances and once again compare our calculations with available experimental data. To our knowledge, no other such studies have been made, particularly for the clusters in the size regimes considered here.

The basic principles involved in both matrix inversion and OS methods for studying the physics of cooperative scattering are outlined in Part I. For completeness, a brief discussion of the OS approach is given here, followed by an outline of the modifications needed for extension of the technique to arbitrarily configured clusters of spheres. The details of the OS method are more easily understood when only two spheres are involved. The total scattered field of the pair can then be decomposed into the different partial fields that result from corresponding orders of scattering between the spheres. These orders of scattering are analogous to the multiple reflections that are set up between the two surfaces of an illuminated thin film. The jth-order partial field of the lth sphere can be expanded in terms of vector spherical harmonics $\mathbf{N}_{mn}$ and $\mathbf{M}_{mn}$ as

$$ {}^{l}\mathbf{E}^{(j)} = \sum_{n=1}^{\infty} \sum_{m=-n}^{n} [{}^{l}a_{mn}^{(j)}\, {}^{l}\mathbf{N}_{mn}^{(3)} + {}^{l}b_{mn}^{(j)} + {}^{l}\mathbf{M}_{mn}^{(3)}], \qquad (1) $$

where the expansion coefficients ${}^{l}a_{mn}^{(j)}$ and ${}^{l}b_{mn}^{(j)}$ constitute, respectively, the jth-order TM and TE modal responses of the lth sphere to the stimulus of the $(j-1)$th-order partial field arriving from the other sphere. If $j = 0$, these quantities correspond to the well-known Mie coefficients. In order to apply the boundary conditions necessary for the explicit determination of the response coefficients, a separate system of coordinates must be associated with the center of each sphere. These coordinate frames are to be related to one another by pure translations. The superscript preceding the vector harmonics in Eq. (1) indicates that the origin of that set of harmonics is located at the center of the lth sphere. The superscript (3) indicates that the radial dependence of those harmonics is based on the spherical Hankel functions of the first kind, whereas the superscript (1) indicates a dependence on the spherical Bessel functions.

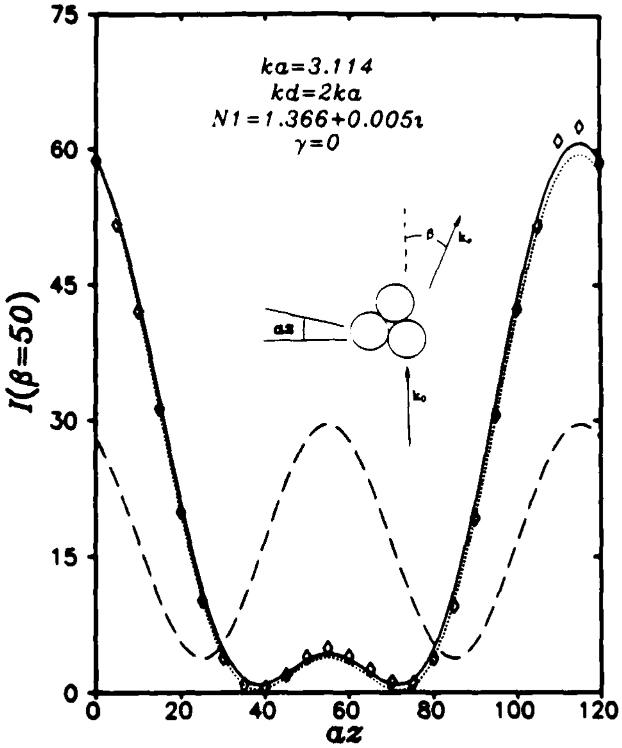

Fig. 1. Intensity of radiation scattered into the angle $\beta = 50°$ by a close-packed triangular cluster of spheres as a function of particle orientation for the particle characteristics and scattering geometry shown. Intensity distributions of this nature are called sparkle functions. The solid curve indicates the converged solution, the dashed curve indicates the case of noninteracting spheres, and the dotted curve indicates the result of first-order interactions between a sphere and a bisphere. The bisphere is illuminated at broadside incidence when $az = 0°$. The diamonds indicate the experimental measurements by R. T. Wang (Space Astronomy Laboratory, University of Florida, Gainesville, Florida).

Other than having to overcome the formidable computational difficulties introduced when dealing with ensembles that are more complicated than the linear chains considered in Part I, one need only replace Eqs. (2) and (4) of that paper with their most general forms,

$$
{}^{i}\mathbf{M}^{(3)}_{mn} = \sum_{\nu=1}^{\infty}\sum_{\mu=-\nu}^{\nu} [{}^{l}\mathbf{M}^{(1)}_{\mu\nu} A^{mn}_{\mu\nu}(k\mathbf{d}_{il}) + {}^{l}\mathbf{N}^{(1)}_{\mu\nu} B^{mn}_{\mu\nu}(k\mathbf{d}_{il})],
$$

$$
{}^{i}\mathbf{N}^{(3)}_{mn} = \sum_{\nu=1}^{\infty}\sum_{\mu=-\nu}^{\nu} [{}^{l}\mathbf{N}^{(1)}_{\mu\nu} A^{mn}_{\mu\nu}(k\mathbf{d}_{il}) + {}^{l}\mathbf{M}^{(1)}_{\mu\nu} B^{mn}_{\mu\nu}(k\mathbf{d}_{il})],
\tag{2}
$$

and

$$
{}^{l}a^{(j)}_{mn} = \sum_{\nu=1}^{\infty}\sum_{\mu=-\nu}^{\nu} [{}^{i}a^{(j-1)}_{\mu\nu} A^{\mu\nu}_{mn}(k\mathbf{d}_{il}) + {}^{i}b^{(j-1)}_{\mu\nu} B^{\mu\nu}_{mn}(k\mathbf{d}_{il})],
$$

$$
{}^{l}b^{(j)}_{mn} = \sum_{\nu=1}^{\infty}\sum_{\mu=-\nu}^{\nu} [{}^{i}a^{(j-1)}_{\mu\nu} B^{\mu\nu}_{mn}(k\mathbf{d}_{il}) + {}^{i}b^{(j-1)}_{\mu\nu} A^{\mu\nu}_{mn}(k\mathbf{d}_{il})],
\tag{3}
$$

respectively. In the equations above, the vector $\mathbf{d}_{il}$ extends from the ith to the lth origin. Equations (2) allow one to express the partial field scattered by any constituent of the cluster in terms of the vector spherical harmonics associated with any other constituent, thereby allowing for an integrable expression for the response coefficients, which ultimately can be cast in the form of Eqs. (3) when only two spheres are involved. The procedure for extending the calculation to three or more spheres is the same as that outlined in Part I. The complete derivation of the scattered fields and a more complete discussion of the translation coefficients $A^{mn}_{\mu\nu}$ and $B^{mn}_{\mu\nu}$ are given in Ref. 4.

Attention should be called to the fact that the index pairs mn and $\mu\nu$ of the translation coefficients in Eqs. (3) are the transpose of those in Eqs. (2). Such a transposition does not appear in Refs. 1 and 4; in this respect, the equations for the jth-order response coefficients that appear in those references are *incorrect.* It must be emphasized that this error is only a typographical one and has no bearing on the outcome of previous calculations.

A comparison of experimental measurements with calculations based on the above theory for the intensity of light (in arbitrary units) scattered into a fixed angle β as a function of particle orientation is presented in Figs. 1 and 2. In Fig. 1, the cluster is a close-packed triangular array of spheres with identical com-

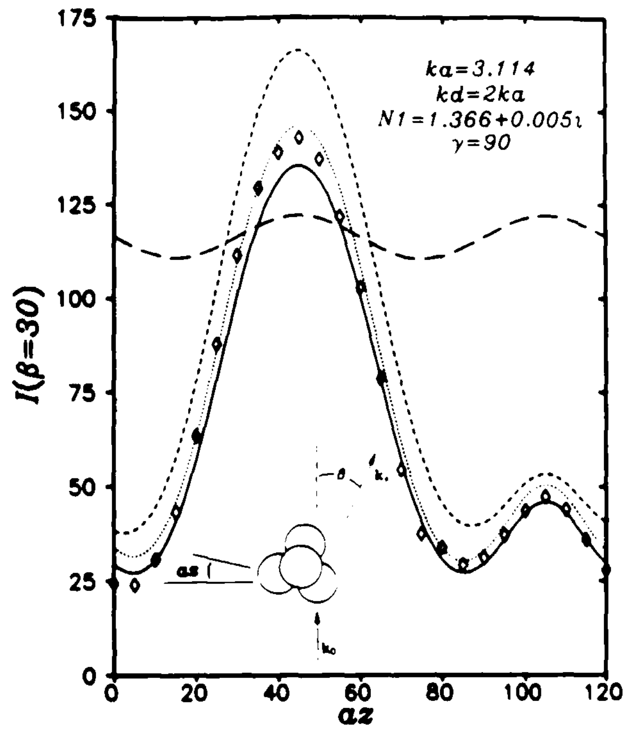

Fig. 2. Sparkle function of a close-packed tetrahedral cluster at $\beta = 30°$. The solid, dotted, short-dashed, and dashed curves correspond to exact, first- plus second-order, first-order, and zeroth-order interactions, respectively. (The scattered fields of noninteracting spheres are produced by zeroth-order multiple scattering.) The diamonds indicate the values measured by R. T. Wang (Space Astronomy Laboratory, University of Florida, Gainesville, Florida).

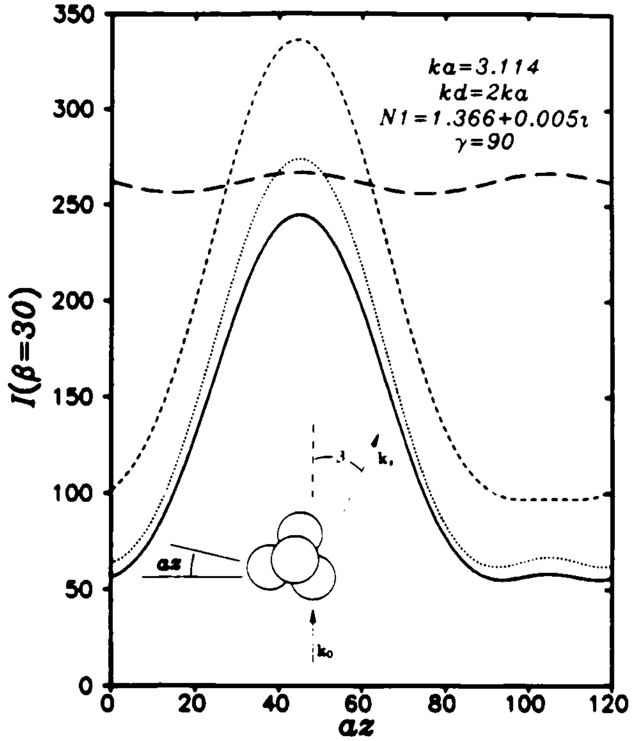

Fig. 3. Same function as Fig. 2, except that the cluster is now a hexahedron, with the fifth sphere located opposite the fourth sphere and invisible from the aspect shown in the inset. (No experimental data are available.)

plex refractive indices $N1$. The incident electric field lies in the plane of the triangle (this state is indicated by the polarization angle $\gamma = 0°$), and the scattering angle is held at $\beta = 50°$. Next, a tetrahedral array is constructed by placing a fourth sphere on top of the triangular array, as shown in the inset of Fig. 2. The polarization angle for this case is taken to be $\gamma = 90°$, i.e., the incident electric field is perpendicular to the plane of the page, and the scattering angle is fixed at $\beta = 30°$. Last, a hexahedral array is studied by placing a fifth sphere on the opposite side of the triangle from the fourth sphere; the scattering parameters are the same as in the case of the tetrahedron, and the results are displayed in Fig. 3. An interesting feature of these results is that the local maximum in $I(\beta = 30°)$ at $az = 105°$, which is about equal to that at $az = 45°$ for the case of a triangular array (not shown), remains almost constant as the fourth and fifth spheres are added, whereas the intensity at $az = 45°$ increases dramatically with each addition. This is evidently caused by constructive and destructive interference between the zeroth and higher scattering orders at $az = 45°$ and $105°$, respectively, although no detailed explanation has been attempted.

The variation of the scattered intensity with the orientation angle az of the particle has been termed the sparkle function. It is interesting to note that as the above clusters are rotated through 120° about their threefold symmetry axis, their sparkle functions display symmetries about two orientations. To see why this is to be expected, it is best to hold the orientation of these clusters fixed at $az = 0°$ and vary the incident angle α of the radiation. At $az = 0°$, the base of the triangle is taken to lie on the x axis of the principal coordinate system. The z axis lies at $az = 90°$. If one takes the reflection of reciprocal (time-reversed) scattering across the yz plane, it can then be seen that $I(\alpha) = I(60° - \alpha + \beta)$. Furthermore, since $I(\alpha) = I(\alpha - 120°)$, one also has $I(\alpha) = I(180° - \alpha + \beta)$. Aside from their intrinsic physical interest, these relations are also useful in determining the degree of uncertainty present in a given set of experimental data, in testing the consistency of a set of calculations, and in expediting such calculations. Similar relations have already been noted in Part I for the case of linear chains.

For the reasons discussed in Ref. 4, the order of the matrices that must be inverted for arbitrarily configured clusters increases dramatically over those encountered when dealing with linear chains. In Part I it was noted that OS calculations tend to run in approximately half the time required by those for matrix inversion, at least for the cases studied thus far. When applied to the close-packed triangular array shown in Fig. 1, OS was found to run fifteen times faster than matrix inversion. The calculations performed for this study could be made much more efficiently if the angle of incidence rather than the orientation of the particle were varied, since each time that az was changed a new set of translation coefficients had to be calculated. The more awkward procedure was preferred for the present investigation, however, because it provided a more rigorous test of the involved routines that are employed in calculating the translation coefficients.

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